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<p>(21) International Application Number: PCT/DK90/00149</p> <p>(22) International Filing Date: 14 June 1990 (14.06.90)</p> <p>(30) Priority data: 3062/89 21 June 1989 (21.06.89) DK</p> <p>(71) Applicant (for all designated States except US): NOVO NORDISK A/S [DK/DK]; Novo Allé, DK-2880 Bagsvaerd (DK).</p> <p>(72) Inventors; and (75) Inventors/Applicants (for US only): FALHOLT, Per [DK/DK]; Lyngbyvej 345B, DK-2820 Gentofte (DK). MARKUSSEN, Erik, Kjaer [DK/DK]; Tornekrogen 18, DK-3500 Vaerloese (DK).</p> <p>(74) Common Representative: NOVO NORDISK A/S; Patent Department, Novo Allé, DK-2880 Bagsvaerd (DK).</p>		<p>(81) Designated States: AT (European patent), AU, BE (European patent), BR, CH (European patent), DE (European patent)*, DK (European patent), ES (European patent), FR (European patent), GB (European patent), IT (European patent), JP, KR, LU (European patent), NL (European patent), SE (European patent), US.</p> <p>Published With international search report.</p>
<p>(54) Title: DETERGENT ADDITIVE GRANULATE AND DETERGENT</p> <p>(57) Abstract</p> <p>The detergent additive granulate is a cocranulate containing at least one enzyme and at least one photobleaching agent. If a detergent, a component of which is such detergent additive granulate, is used for laundering, the photobleaching agent will not tend to adhere selectively to the laundry during the initial washing process.</p>		

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DETERGENT ADDITIVE GRANULATE AND DETERGENT

The concept of incorporation of a photobleaching agent in a detergent is known, reference being made to e.g. DE 3430773, EP 119746, and EP 87833. The photobleaching agent is inactive if not exposed to UV light, and thus, under normal conditions, the photobleaching agent will first be activated and function as a bleaching agent when the clean laundry is dried in the open. This is a clear advantage over the detergents with a normal bleach, which will function as a bleaching agent already in the washing solution and thus damage sensitive detergent additives, especially enzymes.

10 The introduction of photobleaching agents in detergents in solid form is not without problems, though. During the initial stages of the washing process, when the solid detergent agent is introduced into the washing float, particles of pure photobleaching agent or particles of detergent additives, which contains photobleaching agents at the surface thereof, will tend to adhere to the laundry
15 due to a selective adhesion ability of the photobleaching agent to the laundry, and also such photobleaching agent will tend to stay with the laundry without being dissolved during the washing process. Most photobleaching agents possess a very intense color, and such photobleaching agents will create a most undesirable staining of the laundry. In case the photobleaching agent is colorless, the above
20 indicated attachment thereof to the laundry during the washing process will create an uneven concentration of photobleaching agent on the laundry, and in that case an uneven overbleaching action will take place when the laundry is dried in the open.

Thus, there is a need for a detergent additive in solid form
25 comprising a photobleaching agent, whereby during the initial washing process the photobleaching agent will not tend to adhere selectively to the laundry.

Now, according to the invention it has been found that a detergent additive granulate of a special composition will fulfil the above objective.

Thus, the detergent additive granulate according to the invention is a
30 co-granulate containing at least one enzyme and at least one photobleaching agent.

The photobleaching agent can be any photobleaching agent, e.g. Tinolux BBS, vide the following examples, zinc phthalocyanine sulfonate (ZPS), and aluminum phthalocyanine sulfonate (APS).

The detergent additive granulate according to the invention will fulfil
5 the above objective in an especially advantageous manner, if produced by means of a drum granulator, as will later be described in more detail.

The detergent additive granulate can and will usually contain other components besides the components listed above, provided that these components do not damage the enzyme and/or the photobleaching agent.

10 In GB 2.179.053 an aqueous fabric washing detergent is described, which can contain both enzymes and a photobleaching agent. However, this detergent is aqueous, and the above problem, which is solved according to the invention, does not appear at all in relation to aqueous detergents, which exhibit other problems, e.g. stability problems.

15 EP 119.746 describes a detergent powder which contains both photobleach and enzyme. The first step during the production of this washing powder is the formation of the aqueous crutcher slurry, consisting of surfactants, builders, fillers, and a solution of the photobleach. This crutcher slurry is spraydried, whereby the so-called base powder is formed. The enzyme which
20 may optionally be added (vide page 4, line 29 in EP 119.746) has to be added as enzyme containing particles separate from the base powder, due to the fact that the enzyme activity in the base powder would be reduced severely or lost completely, if the enzyme was added to the crutcher solution. Thus, the EP 119.746 contains enzyme free, photobleaching agent containing particles and
25 photobleaching agent free enzyme containing particles. It has been found that the problem of staining and overbleaching is not completely solved in relation to this prior art washing powder.

DE 28 13 198 describes a bleaching agent which contains a special photoactivator. This bleaching agent is produced by cogranulation of all
30 constituents of the bleaching agent to a cogranulate or by granulation of a single constituent on a basis detergent component, whereas according to the invention two sensitive constituents of the detergent are selected, i.e. the enzyme and the photobleaching agent, and cogranulated.

The detergent additive granulate according to the invention can be produced by co-granulation in many ways, e.g. as indicated in the following.

- I. A co-granulate of the so-called T-granulate type, as described in US 4,106,991 (Novo Nordisk A/S, Denmark), i.e. with cellulose fibres.
- 5 II. A co-granulate corresponding to US 4,661,452 (Novo Nordisk A/S, Denmark).
- III. A co-granulate of the core type, as described in DK 4368/88. In this case it is specially preferred that the photobleaching agent is part of the core and that the enzyme is part of the coating.
- 10 IV. A co-granulate of the type described in DK 4369/88. In this case the photobleaching agent is added with the concentrated enzyme solution.
- V. A co-granulate of the so-called "Marumerizer" type, as described in GB 1,362,365.
- VI. A co-granulate of the prill type.
- 15 VII. A co-granulate of the Sparks type, as described in WO 85/05288.
- VIII. A co-granulate of the fluid bed type.

In all cases the photobleaching agent is added as an aqueous solution, preferably as part of the granulation water.

In a preferred embodiment of the detergent additive granulate according to the invention the co-granulate is coated. In this manner the object of the invention is fulfilled to a very high extent. The usual coating agents may be used, e.g. the coating agents described in DK 0121/88, DK 1377/88, and DK 1378/88.

In a preferred embodiment of the detergent additive granulate according to the invention the enzyme is one or more of the enzymes protease, amylase,

lipase, cellulase, oxidase, and amidase. These are the most common enzymes used in the detergent additive.

In a preferred embodiment of the detergent additive granulate according to the invention between 2 and 40% w/w of cellulose fibres are incorporated into the granulate. A granulate of this kind can be produced according to US 4,106,991 (Novo Nordisk A/S, Denmark). In this manner a dust free granulate is obtained which can easily be produced in an industrial scale.

Also, the invention comprises a detergent which contains the detergent additive granulate according to the invention.

10 In a preferred embodiment of the detergent according to the invention the concentration of the detergent additive is between 0.05 and 5% w/w. In this manner the object of the invention is fulfilled, and also, the photobleaching effect is fully satisfactory.

In the following the production of the detergent additive granulate is 15 illustrated in Examples 1 - 11, and the use of the detergent additive granulate is illustrated in Example 12.

EXAMPLE 1

A powder mixture containing a proteolytic enzyme and with the following composition

- 20 8.0 kg of SAVINASE® with an activity of 36.1 KNPU/g
 6.0 kg of fibrous cellulose Arbocel BC 200
 1.6 kg of kaolin, Speswhite ECC1
 2.4 kg of carbohydrate binder
 20.6 kg of finely ground sodium sulphate

25 is transferred to a 130 liter Lödige mixer FM 130 and granulated with 9.7 kg of a granulation liquid made up by mixing of

- 2.0 kg of Tinolux BBS (liquid), Ciba-Geigy
 0.9 kg of carbohydrate binder
 7.0 kg of water

all other conditions during the granulation being identical to Example 1 in US 4.106.991 (Novo Nordisk A/S, Denmark).

KNPU is an activity unit for proteolytic activity, described in the document AF 220/1-GB, KLU is an activity unit for lipolytic activity, described in the document AF 95/5, CSU is an activity unit for cellulolytic activity, described in the document AF 187/3, and KNU is an activity unit for amylolytic activity, described in the document AF 207/1, whereby all the cited documents are available on request from Novo Nordisk, Novo Allé, DK-2880 Bagsvaerd, Denmark.

Tinolux BBS is a photobleaching agent, which is a derivative of 10 tetrabenzo-tetraazaporphine.

The moist granulate is dried on a fluid bed to a water content below 1%, and the sieve fraction of the granulate between 300 μm and 900 μm is taken out as a product. This product is coated with 6.2% of polyethylene glycol (PEG 4000) and 10% of kaolin (Speswhite, ECC) by means of the method described in 15 Example 22 in US 4.106.991 (Novo Nordisk A/S, Denmark).

By means of usual fluid bed techniques, the granulate is further provided with a film coating consisting of 170 ppm of duasyne acid green and 0.25% of hydroxypropyl cellulose.

The final preparation exhibited a proteolytic activity of 5.7 KNPu/g.

20 EXAMPLE 2

A powder mixture containing a lipolytic enzyme and with the following composition

- 0.75 kg of LIPOLASE® with an activity of 860 KLU/g
- 2.25 kg of fibrous cellulose Arbocel BC 200
- 25 1.50 kg of bentonite ECCI ASB350
- 0.90 kg of carbohydrate binder
- 8.90 kg of finely ground sodium sulphate

is transferred to a 50 liter Lödige mixer FM 50 and granulated with 3.4 kg of a granulation liquid made up by mixing of

0.7 kg of Tinolux BBS
0.7 kg of carbohydrate binder
2.0 kg of water

The granulation is carried out as described in Example 1, except for the size of the mixer.

The granulate is dried and sifted as described in Example 1, whereby a granulate with an activity of 31.2 KLU/g is obtained.

EXAMPLE 3

A powder mixture containing a cellulolytic enzyme and with the following composition

3.00 kg of cellulase with an activity of 5340 CSU/g
2.25 kg of fibrous cellulose Arbocel BC 200
0.60 kg of kaolin ECCI Speswhite
0.60 kg of carbohydrate binder
8.85 kg of finely ground sodium sulphate

is transferred to a 50 liter Lödige mixer FM 50 and granulated with 3.4 kg of a granulation liquid made up by mixing of

0.83 kg of Tinolux BBS
0.30 kg of carbohydrate binder
3.00 kg of water

The granulation is carried out as described in Example 1, except for the size of the mixer.

The granulate is dried and sifted as described in Example 1.

EXAMPLE 4

1 kg of dry sifted (300-1000 μm) granulate produced as described in Example 2 is coated in a 5 liter Lödige mixer according to the method described in US 4,106,991 (Novo Nordisk A/S, Denmark), example 22.

5	Prep. 4A	1. coating layer	5.0% PEG 4000
		2. coating layer	10.0% kaolin
		3. coating layer	2.0% PEG 4000
10	Prep. 4B	1. coating layer	5.0% hydrogenated tallow
		2. coating layer	10.0% kaolin
		3. coating layer	2.0% hydrogenated tallow
	Prep. 4C	1. coating layer	5.0% hydrogenated tallow

EXAMPLE 5

1 kg of dry sifted (300-1000 μm) granulate produced as described in Example 3 is coated as described in Example 4.

15	Prep. 5A	1. coating layer	5.0% PEG 4000
		2. coating layer	10.0% kaolin
		3. coating layer	2.0% PEG 4000
20	Prep. 5B	1. coating layer	5.0% hydrogenated tallow
		2. coating layer	10.0% kaolin
		3. coating layer	2.0% hydrogenated tallow
	Prep. 5C	1. coating layer	4.0% hydrogenated tallow

EXAMPLE 6

A powder mixture containing a proteolytic enzyme and with the following composition

- 5 2.7 kg of SAVINASE® with an activity of 48.0 KNPU/g
- 3.0 kg of fibrous cellulose Arbocel BC 200
- 0.8 kg of kaolin ECCI Speswhite
- 1.4 kg of carbohydrate binder
- 11.2 kg of finely ground sodium sulphate

is transferred to a 50 liter Lödige mixer FM 50 and granulated with 5.3 kg of a 10 granulation liquid made up by mixing of

- 3.1 kg of Tinolux BBS
- 0.4 kg of carbohydrate binder
- 0.2 kg of PVP K30
- 1.6 kg of water

- 15 The granulation is carried out as described in Example 1, except for the size of the mixer.

 The granulate is dried and sifted as described in Example 1, whereby a granulate with an activity of 6.0 KNPU/g is obtained.

EXAMPLE 7

- 20 5.0 kg of base granulate produced as described in US 4,106,991 (Novo Nordisk A/S, Denmark), example 1, except that it is based on sodium sulphate as the inert diluent, is provided with a photobleach containing layer by means of a fluidized bed spraying technique, as follows.

 With simultaneous drying in a Glatt fluid bed coater of type WSG5 the 5 25 kg of base granulate is sprayed with an emulsion/suspension consisting of

- 0.75 kg of Tinolux BBS
- 0.50 kg of Bentonite ASB350 (ECCI)
- 0.25 kg of Glascol LS22P (Styrene-acrylic copolymer,
45% microemulsion, Allied Colloids)
- 5 2.00 kg of water,

and with proper selected air inlet and outlet temperature and atomizing air pressure that leaves every single enzyme granulate particle coated with a layer of dry Tinolux and bentonite with Glascol as the binding agent.

EXAMPLE 8

- 10 5 kg of a Tinolux/bentonite coated granulate produced as described in Example 7 is further coated with 100 g of bentonite and 45 g of Glascol LS22P (dry matter) by spraying with an emulsion/suspension consisting of

- 100 g of bentonite ASB 350
- 100 g of Glascol LS22P (45% microemulsion)
- 15 500 g of water

by means of the same fluidized bed coating technique as described in Example 7.

EXAMPLE 9

5.0 kg of base granulate produced as described in US 4,106,991 (Novo Nordisk A/S), example 1, except that it is based on sodium sulphate as the inert
20 diluent, is coated with a photobleach layer by means of a fluidized bed spraying technique.

With simultaneous drying in a Glatt fluid bed coater of type WSG5 the 5 kg of base granulate is sprayed with 0.75 kg Tinolux BBS with proper selected air inlet and outlet temperature and atomizing air pressure that leaves every single
25 enzyme granulate particle coated with a layer of dry Tinolux photobleach.

EXAMPLE 10

5 kg of a Tinolux coated granulate produced as described in Example 9 is further coated with 200 g of bentonite and 135 g of Glascol LS22P (dry matter) by spraying with an emulsion/suspension consisting of

- 5 200 g of bentonite ASB 350
- 300 g of Glascol LS22P (45% microemulsion)
- 1000 g of water

by means of the same fluidized bed coating technique as described in Example 7.

EXAMPLE 11

- 10 A powder composition containing an amylolytic enzyme and with the following composition is prepared for extrusion according to Example 4 in US 4,661,452 (Novo Nordisk A/S, Denmark).

- 0.500 kg of BAN with an activity of 4900 KNU/g
- 0.100 kg of titanium dioxide
- 15 0.275 kg of a carbohydrate binder
- 0.500 kg of calcium carbonate, Sturcal L
- 0.350 kg of anhydrous calcium sulphate
- 0.125 kg of sodium carboxymethyl cellulose, Hercules 7H
- 2.725 kg of finely ground sodium sulphate

- 20 by spraying with 600 g Tinolux BBS containing 100 g of PVP K30 and finally spraying the moist mixture with 200 g of coconut monoethanol amide.

The hot (55°C) moist mixture is extruded using a pellet press type extruder (Simon-Heesen type Monoroll Labor). To obtain an extrudate with a string diameter of 0.8 mm the original Simon-Heesen pellet die with 8 mm holes is

equipped with a screen plate with 0.8 mm holes on the inside, which screen is supported by the die.

After extrusion the extrudate is allowed to cool by blowing cold air through it. After cooling to room temperature it is transferred to the Marumerizer®, which
5 breaks the extrudate to a short sized noodle like product, which is compact and exhibits a well rounded surface. Finally the product is dried in a fluidized bed drier.

The product is directly usable or it may be coated with a protective coating as described in example 22 of US 4,106,991 (Novo Nordisk A/S, Denmark) or in WO 89/08694 (Novo Nordisk A/S, Denmark) or in WO 89/08695
10 (Novo Nordisk A/S, Denmark).

Depending on formulation and extruder type products with particle shapes ranging from pure spheres to short rod like shapes can be produced. Methods for production of spheronized/marumerized products are described in GB 1,362,365 (Novo Nordisk A/S, Denmark) and US 4,661,452 (Novo Nordisk A/S, Denmark).

15 EXAMPLE 12

This example is a spotting example illustrating the use of the detergent additive granulate according to the invention.

Granules from Example 1 are added to a solution of a standard powder detergent in a jar. A piece of white cotton textile is placed on the bottom of the jar.
20 The spotting from the granules on the white textile is evaluated after 1 minute according to the grades shown in the legend.

Dosage of Tinolux 0.02%

Invention	--
Prior art	-
Fluid bed granulate *)	+

5 Dosage of Tinolux 0.04%

Invention	--
Prior art	-
Fluid bed granulate *)	+

Legend:

10 --	absent spotting
-	just detectable spotting
-+	clearly detectable spotting
+	significant spotting
++	very significant spotting

15 *) This prior art fluid bed granulate is produced by addition of the photobleach to a salt carrier in a fluidized bed.

CLAIMS

1. Detergent additive granulate which is a co-granulate containing at least one enzyme and at least one photobleaching agent.
2. Detergent additive granulate according to Claim 1, wherein the co-granulate is 5 coated.
3. Detergent additive granulate according to Claim 1 or 2, wherein the enzyme is one or more of the enzymes protease, amylase, lipase, cellulase, oxidase and amidase.
4. Detergent additive granulate according to Claims 1 to 3, wherein between 10 2 and 40% w/w of cellulose fibres is incorporated into the granulate.
5. Detergent, which contains the detergent additive granulate according to Claims 1 - 4.
6. Detergent according to Claim 5, in which the concentration of the detergent additive granulate is between 0.05 and 5% w/w.

INTERNATIONAL SEARCH REPORT

International Application No PCT/DK 90/00149

I. CLASSIFICATION OF SUBJECT MATTER (If several classification symbols apply, indicate all) ⁶ According to International Patent Classification (IPC) or to both National Classification and IPC IPC5: C 11 D 3/386														
II. FIELDS SEARCHED <div style="text-align: right; margin-right: 100px;">Minimum Documentation Searched⁷</div> <table style="width: 100%; border: none;"> <tr> <td style="width: 25%; border: none;">Classification System</td> <td style="border: none;">Classification Symbols</td> </tr> <tr> <td style="border: 1px solid black; height: 40px; vertical-align: bottom;">IPC5</td> <td style="border: 1px solid black; height: 40px; vertical-align: bottom;">C 11 D</td> </tr> </table> <div style="text-align: center; margin-top: 5px;">Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in Fields Searched⁸</div>			Classification System	Classification Symbols	IPC5	C 11 D								
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IPC5	C 11 D													
SE,DK,FI,NO classes as above														
III. DOCUMENTS CONSIDERED TO BE RELEVANT⁹ <table border="1" style="width: 100%; border-collapse: collapse;"> <thead> <tr> <th style="width: 10%;">Category *</th> <th style="width: 60%;">Citation of Document,¹¹ with Indication, where appropriate, of the relevant passages¹²</th> <th style="width: 30%;">Relevant to Claim No.¹³</th> </tr> </thead> <tbody> <tr> <td style="text-align: center; vertical-align: top;">A</td> <td>EP, A1, 0087833 (UNILEVER NV) 7 September 1983 see claims 11-14, page 9 line 23-27 and examples 5-6, page 18 --</td> <td style="text-align: center; vertical-align: top;">1,3,5</td> </tr> <tr> <td style="text-align: center; vertical-align: top;">A</td> <td>EP, A1, 0119746 (UNILEVER PLC) 26 September 1984, see the whole document --</td> <td style="text-align: center; vertical-align: top;">1,3,5</td> </tr> <tr> <td style="text-align: center; vertical-align: top;">A</td> <td>DE, A1, 2813198 (THE PROCTER & GAMBLE CO.) 19 October 1978, see page 19, lines 1-8 and example 2 on page 26 -- -----</td> <td style="text-align: center; vertical-align: top;">1,3,5</td> </tr> </tbody> </table>			Category *	Citation of Document, ¹¹ with Indication, where appropriate, of the relevant passages ¹²	Relevant to Claim No. ¹³	A	EP, A1, 0087833 (UNILEVER NV) 7 September 1983 see claims 11-14, page 9 line 23-27 and examples 5-6, page 18 --	1,3,5	A	EP, A1, 0119746 (UNILEVER PLC) 26 September 1984, see the whole document --	1,3,5	A	DE, A1, 2813198 (THE PROCTER & GAMBLE CO.) 19 October 1978, see page 19, lines 1-8 and example 2 on page 26 -- -----	1,3,5
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<div style="display: flex; justify-content: space-between;"> <div style="width: 45%;"> <p>* Special categories of cited documents:¹⁰</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> </div> <div style="width: 45%;"> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance, the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p>"Y" document of particular relevance, the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.</p> <p>"&" document member of the same patent family</p> </div> </div>														
IV. CERTIFICATION <table style="width: 100%; border: none;"> <tr> <td style="width: 50%; border: none;"> Date of the Actual Completion of the International Search 26th September 1990 </td> <td style="width: 50%; border: none;"> Date of Mailing of this International Search Report 1990-09-26 </td> </tr> <tr> <td style="border: none;"> International Searching Authority <div style="text-align: center;">SWEDISH PATENT OFFICE</div> </td> <td style="border: none;"> Signature of Authorized Officer <div style="text-align: center;"><i>Dagmar Järvman</i> Dagmar Järvman</div> </td> </tr> </table>			Date of the Actual Completion of the International Search 26th September 1990	Date of Mailing of this International Search Report 1990-09-26	International Searching Authority <div style="text-align: center;">SWEDISH PATENT OFFICE</div>	Signature of Authorized Officer <div style="text-align: center;"><i>Dagmar Järvman</i> Dagmar Järvman</div>								
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International Searching Authority <div style="text-align: center;">SWEDISH PATENT OFFICE</div>	Signature of Authorized Officer <div style="text-align: center;"><i>Dagmar Järvman</i> Dagmar Järvman</div>													

**ANNEX TO THE INTERNATIONAL SEARCH REPORT
ON INTERNATIONAL PATENT APPLICATION NO.PCT/DK 90/00149**

This annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report.
The members are as contained in the Swedish Patent Office EDP file on **90-08-28**
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Patent document cited in search report	Publication date	Patent family member(s)	Publication date	
EP-A1- 0087833	84-09-07	AU-B-	544554	85-06-06
		AU-D-	1143383	83-08-25
		CA-A-	1202452	86-04-01
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